

Electrostatic Tuning of the Properties of Disordered Indium Oxide Films near the Superconductor-Insulator Transition

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Abstract

The evolution with carrier concentration of the electrical properties of amorphous indium oxide (InO) thin films has been studied using electronic double layer transistor configurations. Carrier variations of up to $7 \times 10^{14} \text{ carriers/cm}^2$ were achieved using an ionic liquid as a gate dielectric. The superconductor-insulator transition was traversed and the magnitude and position of the large magnetoresistance peak found in the insulating regime were modified. The systematic variation of the magnetoresistance peak with charge concentration was found to be qualitatively consistent with a simulation based on a model involving granularity.

The competition between disorder and superconductivity has long been the subject of theoretical and experimental study[1–6]. By varying the nominal disorder a superconductor-insulator (SI) transition is found in materials such as amorphous Bi (*a*-Bi)[7], TiN_x[8], and InO_x[9, 10]. Also Parendo *et al.* electrostatically tuned the SI transition of *a*-Bi using SrTiO₃ as both a substrate and a gate dielectric[11]. Here we focus on the electrostatic tuning of amorphous InO_x which we will refer to as InO. This material is of interest because of the novel feature of a giant magnetoresistance (MR) peak at low temperatures in the insulating regime, as first reported by Paalanen, Hebard, and Ruel[12], and subsequently studied by many workers[10, 13]. Direct evidence of localized Cooper pairs, both above the transition temperature, and in the insulating regime has been reported in scanning tunneling microscope (STM)[14] and planar tunneling junction experiments[15], where the existence of a superconducting gap was inferred from the form of the density of states. Recent theoretical works[16–18] suggest that the non-monotonic behavior of the MR of disordered systems in the insulating state may be associated with a competition between the single-grain charging energy E_c and the electron pairing energy Δ , modeling the film as a granular superconductor. Despite many experimental and theoretical works, little is known about the role of carrier density in the SI transition of InO and the effect of carrier density variations on the MR peak. Here we present data relating to these issues and suggest a qualitative explanation of the results based on a numerical simulation using one of the models[18].

Amorphous InO films were grown on $0.6 \times 0.6 \text{ cm}^2$ SiO substrates and were patterned during growth using a physical (shadow) mask. Sample thicknesses ranged from 10 nm to 6 nm. The 6 nm samples were all insulating as grown and are the central focus of this work. Gold electrodes, 10 nm thick, were deposited on the InO films. An ionic liquid (IL), DEME-TFSI, was used as a gate dielectric with a Pt coil top gate to form an electric double

layer transistor (EDLT). A schematic of the top-gated EDLT can be found in our previous work[19]. (Earlier, Misra, McCarthy, and Hebard gated InO films with ILs but did not explore their superconductivity[20].)

The sheet resistances R_s of the films were determined employing a four-probe electrode configuration. A ³He refrigerator with a superconducting magnet enabled us to vary the temperature T between 300K and 0.40K and the magnetic field H between 0T and 9T. Gate voltages V_g , ranging from +3V to -3V were used to induce or deplete charge carriers, which are electrons. The various values of V_g were applied at a temperature of 240K and were held constant throughout the subsequent cooling and measurement.

The T dependencies of R_s , of two InO samples at different values of V_g are plotted in Fig. 1. Both samples were grown under the same conditions *i.e.*, at the same oxygen pressure and at the same time. However their initial resistances were adjusted so as to be different, by annealing at 65°C under vacuum for different periods of time. Both samples exhibited large changes in R_s upon gating. Unfortunately, Hall effect measurements of such highly disordered systems were not possible, except in the case of a few other samples (not shown) that had relatively low normal state resistances. The process of gating was found to be reversible, with minor hysteresis. This suggests that apart from the remote possibility of a reversible chemical reaction, the gating process is electrostatic. To determine and/or confirm the carrier modulation of the system, we used an electrochemical technique known as the Chronocoulometry[21]. Using this technique, we observed changes in the sheet (2D) carrier density or charge transfer (Δn_{sheet}) of up to $7 \times 10^{14} \text{ carriers/cm}^2$. However we did not characterize the charge transfer at each gate voltage for the films reported here.

The accessible charge transfer was large enough for us to observe the SI transition of InO as shown in Figs. 1(a) and 1(b). The right hand panels of Fig. 1 show R_s vs. T behavior of the insulating state; both Arrhenius

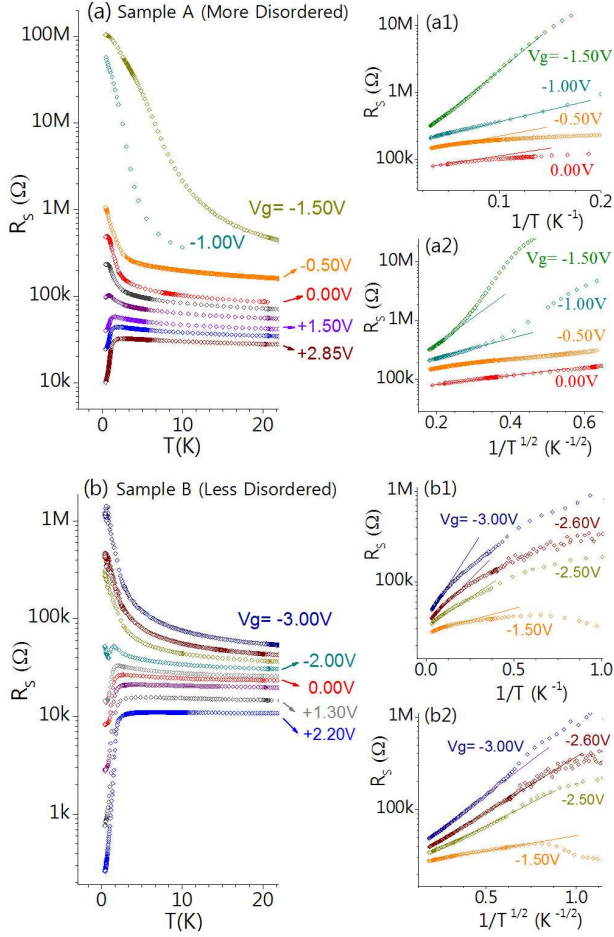


Figure 1: (Color online) The temperature dependence of R_s at various V_g of samples A (a) and B (b). The onset of superconductivity (the initial downward dip in the R_s vs. T curve) occurs at 0.85K, with $V_g = +1.00$ V for sample A, and at 1.47K, with $V_g = -2.00$ V for sample B respectively. On the right hand panels, both R_s vs. $1/T$ and $1/T^{1/2}$ are plotted for sample A (a1-a2) and for sample B (b1-b2). Note the cross-over from Arrhenius to ES VRH at $V_g = -0.50$ V shown in (a1-a2), whereas (b1-b2) shows ES VRH over all ranges of V_g .

$R = R_0 \exp(T_0/T)$ and Efros-Shklovskii variable range hopping (ES VRH) $R = R_0 \exp(T_0/T)^{1/2}$ were observed. Comparing Figs. 1(a1) and 1(b1), the more insulating sample shows a wider range of Arrhenius activated transport, suggesting that it has a hard gap in its single-particle density of states. A cross-over from Arrhenius to ES hopping is observed as the film becomes less resistive with increasing carrier concentration. The observed ES hopping suggests the presence of a soft gap (Coulomb gap) in the density of states near the Fermi level, which is due to long range interactions between electrons in the system[22]. This has also been reported in other granular systems[8, 23]. Both samples A and B exhibited SI transitions tuned by carrier modulation, which is clearer

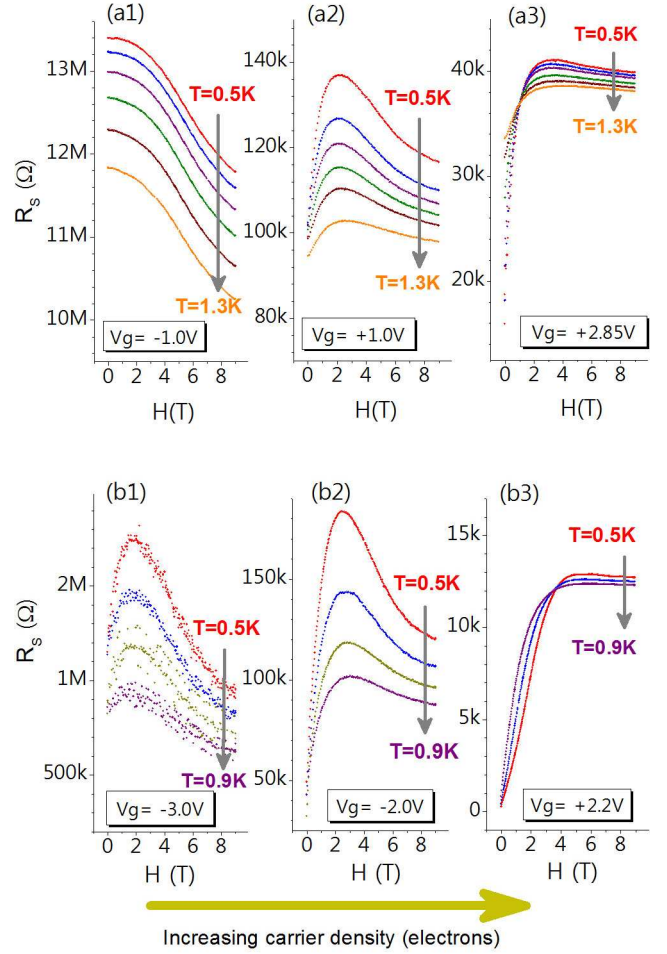


Figure 2: (Color online) R_s vs. H at various values of T and V_g for sample A, labeled (a1-a3) and sample B, labeled (b1-b3). The density of electrons increases from the left to the right. Each curve represents a separate isotherm of R_s vs. H , where T ranges from 0.5~1.3K for sample A (more disordered, top) and from 0.5~0.9K for sample B (less disordered, bottom).

in the case of sample B [Fig. 1(b)].

The MR measurements of these two samples at different gate voltages are shown in Fig. 2. As mentioned, such MR peaks in disordered systems have been reported in a variety of experiments[2, 10, 12, 13]. The most significant feature of the data for sample A [Figs. 2(a1), 2(a2), and 2(a3)] is the transition from negative MR to positive MR followed by downward slope of R_s (MR peak) upon further increase of H , all taken at fixed T over the range from 0.5K to 1K.

We have attempted to quantify some of our observations of the MR peak. A magnetic length ζ associated with the field at the peak H_{peak} , can be estimated as $\zeta = (\Phi_0/H_{peak})^{1/2}$, where $\Phi_0 = 2.07 \times 10^{-15} \text{ T} \cdot \text{m}^2$ is the flux quantum. Values of the peak magnetic field, H_{peak}

at different gate voltages, all taken from the isotherm curves of R_s vs. H at $T = 0.5K$, were found to be monotonically increasing functions of the gate voltage. This implies that ζ decreases with increasing carrier density. A measure of the size of the MR peak can be taken to be $R_{norm} = R_s(H_{peak})/R_s(H = 2H_{peak})$. Figure 3 shows a plot of R_{norm} vs. ζ for each of the samples, where different points correspond to different values of gate voltage. The inset to Fig. 3 shows a plot of H_{peak} vs. the reciprocal of R_s at 100K. The latter is proportional to the carrier concentration assuming a Drude resistance at high temperatures and assuming that the gate voltage changes the carrier concentration and not the disorder[24]. Note that in the inset, curves of H_{peak} vs. $1/R_{s,100K}$ of the two samples collapse onto each other, suggesting a possible universal behavior.

There is an apparent connection of this behavior to sample morphology. Atomic force microscope (AFM) images (Fig. 3, bottom, left) of these films exhibited roughness which could be interpreted as granularity, even though the films are completely connected. A Power Spectral Density (PSD) analysis (Fig. 3, bottom, right) of the AFM images revealed a structural length scale implying a weak level of correlation in the morphology. These lengths are indicated by the vertical dotted lines with estimated error bars for the two curves of Fig. 3. Note that the lengths corresponding to the PSD peaks are close to the values of ζ corresponding to the maxima of R_{norm} vs. ζ especially in the case of sample A. Thus when the magnetic length scale corresponds to the structural length, there is a maximum in the strength of the MR peak.

A granular morphology may not be essential for a system to exhibit granular like behavior. The oxygen concentration may be the key to determining an effective granularity in the amorphous InO system. A recent experimental work[25] showed that mesoscale spatial variations in the oxygen concentration are present in nominally homogeneous, amorphous InO films. Such chemical inhomogeneity can induce local carrier density fluctuations resulting in mesoscale inhomogeneity of the pairing energy Δ . However, we don't know if the mesoscale thickness variations in the films of the present work are correlated with variations in the local indium to oxygen ratio.

In a disorder-driven SI transition, the spatial inhomogeneity of the pairing energy Δ can be very important. In the insulating regime, in some models, the system may break up into superconducting islands, *i.e.*, become effectively granular[5]. In this picture global superconductivity occurs when there is phase coherence between the localized order parameters of different grains, while the energy to break a pair is much larger and remains nonzero even when the coherence (and thus the global superconductivity) is destroyed[13]. In Ref. [6] such a spatial fluctuation of the order parameter amplitude and

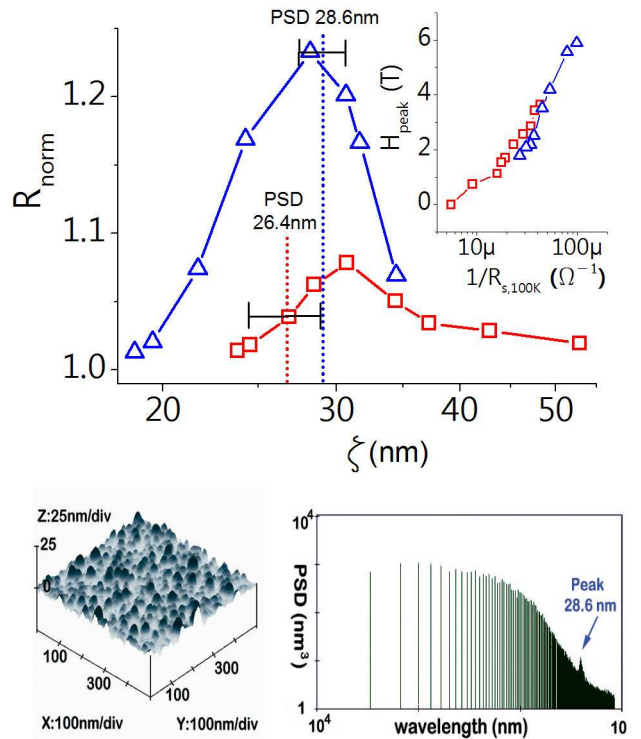


Figure 3: (Color online) (Top) The plot of R_{norm} vs. ζ for samples A (red, square) and B (blue, triangle). The inset shows the values of peak field H_{peak} where the maximum of the MR at $T = 0.5K$ is found as a function of $1/R_{s,100K}$ in zero magnetic field, which is proportional to the carrier concentration. The quantity defined as R_{norm} is a measure of the magnitude of the MR peak. The maximum R_{norm} occurs very near the length extracted from the PSD of the AFM surface profile (dotted vertical lines) for sample A and very close to that length for sample B. (Bottom) The AFM image (left) and the PSD analysis (right) showing a peak, corresponding to the periodicity of surface morphology, for sample B. Similar results were observed for sample A.

corresponding phase correlation have been demonstrated using a negative U-Hubbard model. Near the SI transition, it has been suggested[26, 27] that the MR peaks in disordered systems arise because magnetic fields affect the concentration and size of superconducting islands, so that as these islands shrink with increasing field there is a transition from Cooper pair-dominated to single electron-dominated transport.

On the other hand, reduction of the superconducting pairing energy Δ within grains can itself lead to a tradeoff between conduction by Cooper pairs and conduction by unpaired electrons, and thus (potentially) to a MR peak, even when the concentration and the size of the superconducting islands are fixed. Recent theoretical works[17, 18] study a model with fixed size and concentration of superconducting grains, presumably set by the film morphology, and show how a MR peak deep in the insulating state can arise as a result of the reduction of

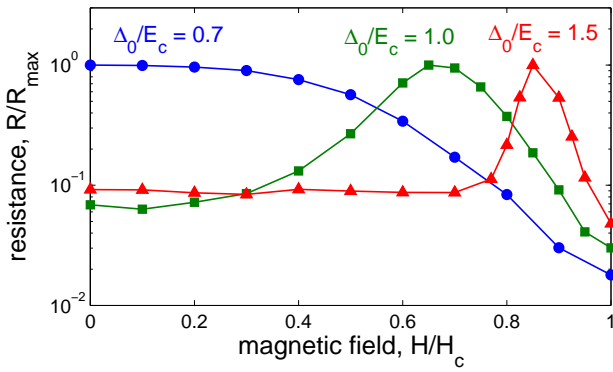


Figure 4: (Color online) Simulation of the resistance of a 2D array of identical superconducting grains deep in the insulating state as a function of H . Different curves are labeled with their corresponding values of Δ_0/E_c , and are normalized to their maximum R_{max} . As Δ_0 is increased, which presumably corresponds to larger carrier density, a MR peak develops that shifts to larger magnetic field, in qualitative agreement with what is seen in Fig. 2. Here all curves correspond to a temperature such that $k_B T = 0.1 E_c$ and have localization lengths ξ_1 and ξ_2 for single-electron and pair conductivity, respectively, satisfying $\xi_2/\xi_1 = 8$.

the superconducting gap Δ with increased H . This predicts that near the MR peak and at low temperature the conduction should be described by ES VRH, as shown in Figs. 1(a2) and 1(b2). Both approaches lead to an insulator in which Cooper pairs with nonzero Δ are formed in the insulating regime of the system and are responsible for the MR peak.

The shift of the MR peak to higher magnetic fields with increasing carrier concentration, as shown in Figs. 2 and 3, can be explained qualitatively within the context of the theory of Ref. [16–18]. Increasing the carrier density presumably increases the density of states at the Fermi level within the superconducting grains, thereby driving up the zero-field superconducting gap, Δ_0 . A larger Δ_0 implies that a larger H is required in order to reduce Δ to the value of the grain charging energy E_c , so that the MR peak shifts to higher H . In this way the transition from negative MR [as in Fig. 2(a1)] to a peak at an intermediate H [Fig. 2(a2)] to a peak at a large H [Fig. 2(a3)] can be understood.

As an example, Fig. 4 shows values of the resistance of a simulated 2D array of regularly-spaced, monodispersed superconducting grains as a function of H , calculated using the method described in Ref. [18]. At small Δ_0/E_c , the conductivity is primarily due to hopping of unpaired electrons, and there is a monotonic negative MR [as seen, for example, in Fig. 2(a1)]. At larger Δ_0/E_c , which ostensibly corresponds to larger carrier density, the MR develops a peak associated with a trade-off between conductivity by single electrons and conductivity by Cooper pairs. This peak moves to larger H as Δ_0/E_c is increased

[as in Fig. 2]. For the simulation of Fig. 4 we have assumed a conventional BCS-like dependence of Δ on the field H : $\Delta = \Delta_0 \sqrt{1 - (H/H_c)^2}$. In this way the data shown in Fig. 2 is consistent with the concept of tuning the local superconducting gap by modulating the carrier density. Unfortunately, the simulation method used to generate Fig. 4 cannot be used for a quantitative determination of the relationship $\Delta(n)$, since this requires a knowledge of the H -dependence of the gap as well as the relative localization lengths ξ_1 and ξ_2 for unpaired and paired electron hopping. We also caution that the simulation technique is applicable only for the heavily-insulating limit, and in this sense our comparison between Figs. 2 and 4 is only qualitative. It should also be noted that within this simple model a strong MR peak develops only at relatively large ξ_2/ξ_1 . A final caveat is the possibility that other models may give similar results.

In conclusion, we have demonstrated the evolution of the MR peak found in InO as a function of carrier density. The transition from a negative MR to a strong MR peak followed by the suppression of the MR peak was observed near the SI transition tuned by carrier modulation. Our findings support the presence of localized Cooper pairs in the insulating regime and are qualitatively consistent with models based on granularity of the system. The reason the amplitude of the MR peak is a maximum when the magnetic length is close to a length scale associated with disorder in the films remains an open question.

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